

# Giant Faraday rotation in single- and multilayer graphene

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Optical Faraday rotation is one of the most direct and practically important manifestations of magnetically broken time-reversal symmetry. The rotation angle is proportional to the distance traveled by the light, and up to now sizeable effects were observed only in macroscopically thick samples and in two-dimensional electron gases with effective thicknesses of several nanometers. Here we demonstrate that a *single atomic* layer of carbon - graphene - turns the polarization by several degrees in modest magnetic fields. The rotation is found to be strongly enhanced by resonances originating from the cyclotron effect in the classical regime and the inter-Landau-level transitions in the quantum regime. Combined with the possibility of ambipolar doping, this opens pathways to use graphene in fast tunable ultrathin infrared magneto-optical devices.

Graphene's unusual physical properties<sup>1,2</sup> make it highly attractive for both fundamental research and novel applications. Importantly, the conduction and valence bands in graphene show a relativistic massless dispersion and a chiral character of the electronic wavefunctions, resulting in non-equidistant Landau levels (LLs) ( $n = 0, \pm 1, \pm 2 \dots$ ):

$$E_n = \text{sign}(n) \sqrt{2e\hbar v_F^2 |nB|} \quad (1)$$

in a perpendicular magnetic field  $B$ , which includes an anomalous LL at zero energy  $E_0 = 0$  ( $e > 0$  is the elementary charge and  $v_F \approx 10^6$  m/s is the Fermi velocity). As a consequence, graphene shows the half-integer quantum Hall effect<sup>3,4</sup>, first observed in exfoliated graphene flakes<sup>5,6</sup> and later, in graphene epitaxially grown on SiC<sup>7-9</sup>.

The optical properties of graphene are equally unusual. The relativistic and gapless dispersion results in a universal and constant optical conductivity of  $\sigma_0 = e^2/4\hbar$ <sup>4,10-15</sup> for the photon energies  $\hbar\omega$  above twice the Fermi en-

ergy ( $\epsilon_F$ ). In magnetic field, the non-equidistant spacing of the LLs gives rise to a spectacular series of absorption peaks, corresponding to transitions between various levels<sup>16-19</sup>. In contrast, only one peak, centered at the cyclotron frequency  $\omega_c = eB/m_c$ , where  $m_c$  is the cyclotron mass, is observed in conventional 2D electron gases (2DEGs), produced in semiconductor heterostructures.

Up to now, experimental magneto-optical studies of graphene<sup>16,17,19</sup> were focused on the diagonal conductivity component  $\sigma_{xx}(\omega, B)$ ; the off-diagonal, or AC Hall<sup>20</sup>, conductivity  $\sigma_{xy}(\omega, B)$  was addressed however only theoretically<sup>18,21,22</sup>. The diagonal and Hall conductivities are directly related to the absorption  $1 - T$  and Faraday rotation angle  $\theta$  (Fig.1) respectively. Adopting the thin-film approximation and keeping only the terms linear in conductivity<sup>16,21</sup> the following expressions are obtained:

$$1 - T(\omega, B) \approx 2Z_0 f_s(\omega) \text{Re}[\sigma_{xx}(\omega, B)] \quad (2)$$

$$\theta(\omega, B) \approx Z_0 f_s(\omega) \text{Re}[\sigma_{xy}(\omega, B)] \quad (3)$$

where  $T$  is the substrate-normalized transmission (see the Methods section),  $Z_0 \approx 377 \Omega$  is the impedance of vacuum and  $f_s(\omega)$  is a spectrally featureless function specific to the substrate (as described in the Methods section). Notably, the diagonal conductivity is formally independent of the sign of the charge carriers, while the DC Hall conductivity, which is sensitive to the carrier type, lacks spectral information. Therefore, studying the Faraday rotation is needed to complete the picture of the dynamics of electrons and holes in graphene.

Graphene, epitaxially grown on silicon carbide, is well adapted for magneto-optical studies because of its well-controlled morphology and essentially unlimited size<sup>23,24</sup>. Here, we used single and multi layer graphene (SLG and MLG), grown respectively on the Si-terminated<sup>24</sup> and C-terminated<sup>23</sup> surfaces of 6H-SiC. The first sample underwent H-passivation of the Si dangling bonds<sup>25,26</sup>, resulting in quasi-free standing SLG. The MLG consists of 4 -

6 rotated atomic layers (see the Methods section). These samples allowed us to explore the cases of both electron and hole doping as well as to access both the classical (high doping) and quantum (low doping) regimes as described below.

Figure 2a shows the Faraday angle  $\theta$  measured on SLG at 5 K in magnetic fields up to 7 T in the far-infrared range as described in the Methods section. The spectra show a strongly field dependent edge-like structure, giving rise to a positive rotation at low energies and negative  $\theta$  at high energies. The maximum Faraday rotation exceeds 0.1 radians ( $\sim 6$  degrees), which is an exceptionally large effect given that it comes from a single layer. Measurements on the bare substrate did not reveal any Faraday effect, hence the observed rotation comes exclusively from the carbon monolayer. The inset shows the field dependence of  $\theta$  at 10 and 27 meV. The curves follow an approximately linear dependence, with opposite slopes of  $+18.5$  mrad/T and  $-4.5$  mrad/T respectively.

The zero-field normalized transmission spectra  $T(B)/T(0)$  of SLG also reveal a strong magnetic field dependence (Fig. 2b). The inset shows the absorption  $(1 - T)$  at 0 and 7 T. One can clearly see a strong Drude peak, signaling a high doping level. The peak center shifts from zero to a finite energy at 7 T, which is due to the cyclotron resonance as we discuss below. Although an estimate of the doping can be made from the integrated intensity of the Drude peak, the most direct measurement of the Fermi energy comes from the absorption spectra in the mid-infrared range (Fig. 2c), where an absorption edge is easily recognized. This is expected to be at  $2|\epsilon_F|$  due to Pauli blocking<sup>4,10,11,14,15</sup>, giving an estimate for  $|\epsilon_F| = 0.34 \pm 0.01$  eV.

Independently, we have performed angle resolved photoemission spectroscopy (ARPES) on a similar sample (Fig. 2d). The obtained dispersion curves show that the sample is p-doped (the Fermi level is in the valence band). The Fermi energy inferred from optical measurements is in a reasonable agreement with an extrapolation of the occupied bands measured by ARPES. Below we show that the carrier type can also be directly extracted from the Faraday rotation.

The doping level and field strength used here puts the system in the classical regime, where the separation between the LLs at the Fermi energy is much smaller than  $\epsilon_F$ <sup>15,19</sup>. In this case, the Dirac quasiparticles are expected to exhibit the classical cyclotron resonance effect. Indeed, the data in Figures 2a and 2b at each separate field are well fitted (dashed lines) using equations (2) and (3) and the classical Drude formulas:

$$\sigma_{xx}(\omega, B) = \frac{2D}{\pi} \cdot \frac{1/\tau - i\omega}{\omega_c^2 - (\omega + i/\tau)^2} \quad (4)$$

$$\sigma_{xy}(\omega, B) = -\frac{2D}{\pi} \cdot \frac{\omega_c}{\omega_c^2 - (\omega + i/\tau)^2} \quad (5)$$

where  $D$  is the Drude weight,  $\omega_c$  is the cyclotron frequency (which is positive for electrons and negative for

holes) and  $\tau$  is the scattering time. From here we see that the observed large Faraday rotation is associated with the cyclotron resonance. Furthermore, the curve  $\theta(\omega)$  reveals the value and sign of the cyclotron frequency, since  $|\omega_c|$  coincides with the position of the maximum absolute slope  $|d\theta(\omega)/d\omega|$  (shown by the arrow in Figure 2a for 7 T), while the sign of the slope matches the sign of  $\omega_c$ . In our case, the negative slope is an unmistakable signature of hole doping.

In the picture of non-interacting Dirac fermions, the Drude weight (at zero magnetic field) is equal to  $2\sigma_0|\epsilon_F|/\hbar$ . Using this relation, we estimate  $|\epsilon_F| = 0.35 \pm 0.03$  eV from the Drude weight extracted from the fit. This agrees with the estimate based on the mid-infrared absorption threshold but is subject to relatively large error bars.

The cyclotron frequency (Fig. 2e) demonstrates an approximately linear growth with field, in agreement with the theoretical relation<sup>10,15</sup>

$$\omega_c = \frac{eBv_F^2}{\epsilon_F}. \quad (6)$$

However, the theoretical curve using the value  $\epsilon_F = -0.34$  eV and  $v_F = 1.02 \times 10^6$  m/s extracted from the mid-infrared optical spectra and ARPES data respectively (dashed line in Fig. 2e) is somewhat lower than the experiment. Although the reason of this deviation is currently unknown, one can speculate about its possible relation to many-body effects. Interestingly an electron-plasmon coupling was recently observed in similar samples<sup>27</sup>.

The carrier mobility  $\mu$  can also be extracted using the relation  $|\omega_c|\tau = \mu|B|$  (Fig. 2f). The mobility varies weakly with field between  $2,400$ - $3,000$  cm<sup>2</sup>/(V·s). Even at 7 T,  $\mu|B| \sim 1$ , which means that the cyclotron resonance is significantly damped due to disorder. This can also be seen from the large width ( $1/\tau \sim 10$  meV) of the cyclotron peak in the inset of Figure 2b.

The Faraday rotation and absorption observed in multi layer graphene is strikingly different (Figure 3). Both the Faraday angle and absorption spectra show additional, strongly field dependent resonance structures, marked by arrows. As was found in previous studies<sup>16,17</sup>, they correspond to optical transitions between individual LLs. The series of transitions with energies  $E_1 - E_0$ ,  $E_2 - E_1$  and  $E_3 - E_2$  can be identified and fitted using equation (1), which gives  $v_F = 1.00 \pm 0.01 \times 10^6$  m/s (inset of Fig. 3b). Although inter-LL absorption peaks have been observed before<sup>16,17</sup>, the measurement of the Faraday rotation at the same conditions is the key novelty of the present work. Now we can distinguish transitions involving electrons from the ones involving holes. In particular, the positive sign of  $d\theta(\omega)/d\omega$  at the transition energies, which coincide with the inflection points, unequivocally determines that the transitions are between electron bands. The low frequency part of the spectra features a cyclotron-resonance like structure, similar to the data on SLG. The sign of the Faraday angle now

corresponds to electron doping.

Since only transitions between occupied and empty levels are allowed, the simultaneous presence of  $0 \rightarrow 1$ ,  $1 \rightarrow 2$  and  $2 \rightarrow 3$  transitions and the low-frequency cyclotron structure in MLG arises from a variation of the Fermi energy across different layers. The layer closest to the substrate is highly doped<sup>28</sup>; the doping in subsequent layers decreases exponentially with layer number. The most strongly doped innermost layer gives rise to the cyclotron resonance, as in SLG. Meanwhile, the individual inter-LL transitions originate from the weakly doped layers, which are in the quantum regime, as can be seen from the square-root field dependence of the transition energies. The maximum Faraday angle in this sample is smaller than in the monolayer sample, because the innermost layer in MLG is more weakly doped than the latter.

Summarizing the data on both samples, we point out that the Faraday rotation is a powerful contact-free tool to distinguish the carrier type and measure the mobility in graphene. Both in the classical (high doping) and quantum (low doping) limits this technique reveals magneto-optical resonances due to either the cyclotron effect or to inter-LL transitions. The technique has the potential to distinguish electronic contributions from separate graphene layers. However, our most striking observation is the giant value of the rotation ( $> 0.1$  rad for a single atomic layer). This is much larger than the fine-structure constant ( $\sim 10^{-2}$ ) - the predicted scale for the Faraday angle, associated with the quantized Hall conductance<sup>21,22,29,30</sup>. We demonstrated that this enhancement is due to the proximity to the resonance frequency. Although in 2DEGs the cyclotron resonance gives rise to comparable absolute rotations<sup>30,31</sup>, the rotation comes from an effective layer which is about one order of magnitude thicker than graphene. Furthermore, while in 2DEGs the cyclotron frequency is doping independent, in graphene it can be tuned with doping (electrostatic or chemical) as seen from Equation (6), and much higher frequencies can be achieved at the same fields.

The Faraday effect and the associated magneto-optical Kerr effect are widely used in optical communication, data storage and computing. We suggest that the use of graphene for fast tunable ultrathin magneto-optical devices should be explored. Indeed, the possibility of easy and fast ambipolar doping together with a strong Faraday effect in a wide frequency range is a unique combination not present in other known materials.

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## I. METHODS

### A. Samples

Single and multi layer epitaxial graphene were made by graphitization of the surface of semi-insulating 6H-SiC substrates in an argon atmosphere<sup>24</sup>. The sample size was  $10 \times 10$  mm<sup>2</sup>. SLG was obtained on the Si terminated face of SiC by first preparing a  $(6\sqrt{3} \times 6\sqrt{3})$  reconstructed surface (the so-called buffer layer) by annealing in 1 bar of Ar at a temperature of 1450 °C, which was subsequently converted into quasi-freestanding graphene by intercalation of hydrogen<sup>25,26</sup>. MLG was grown on the C-terminated face of SiC. In either case, the back side of the substrate was cleaned from undesirably grown graphene using scotch-tape and was checked to be graphene free using X-ray photo emission (XPS). Likewise, the layer thicknesses of the SLG and MLG samples were determined using XPS measurements<sup>32</sup>.

### B. Magneto-optical experiment

The sample was mounted in a split-coil superconducting magnet attached to a Fourier-transform spectrometer. A global light source and He-cooled bolometer detector were used. Two grid-wire gold polarizers (one fixed and one rotating) were mounted before and after the sample. The Faraday rotation was deduced from the angle of the rotating polarizer at which the intensity was at minimum. The substrate-normalized transmission  $T$  was measured with one polarizer only and is defined as the ratio between the intensity of the light transmitted through the graphene on SiC and through bare substrate.

In the thin-film limit, the absorption and the Faraday angle are related to the diagonal and the Hall conductivity respectively by equations (2) and (3), where we made use of a spectrally smooth dimensionless function  $f_s(\omega)$ , which depends on the refractive index  $n_s(\omega)$  and the extinction coefficient  $k_s(\omega)$  and the thickness  $d$  of the substrate. In our measurements, the Fabry-Perot interference in the substrate was not spectrally resolved. In this case:

$$f_s(\omega) = \left( \frac{1}{n_s + 1} + \frac{2n_s}{n_s^2 - 1} \cdot \frac{q^2}{1 - q^2} \right) \quad (7)$$

where  $q = [(n_s - 1)/(n_s + 1)]^2 \exp[-(2\omega/c)k_s d]$ .

Note that for a weakly absorbing substrate  $q \approx [(n_s - 1)/(n_s + 1)]^2$  and therefore  $f_s \approx (n_s^2 + 3)/(4n_s^2 + 4)$  while for a strongly absorbing substrate  $q \ll 1$  and  $f_s \approx 1/(n_s + 1)$ . Experimentally, both  $n_s$  and  $k_s$  can be determined from the measurement of the absolute transmission and reflection spectra of the bare substrate. Figure IB shows the function  $f_s$  of the used SiC substrate

( $d \approx 370$  microns) at 5 K. Below 60 meV, this function is

constant; at higher photon energies it decreases because of the presence of a strong optically active phonon in SiC.

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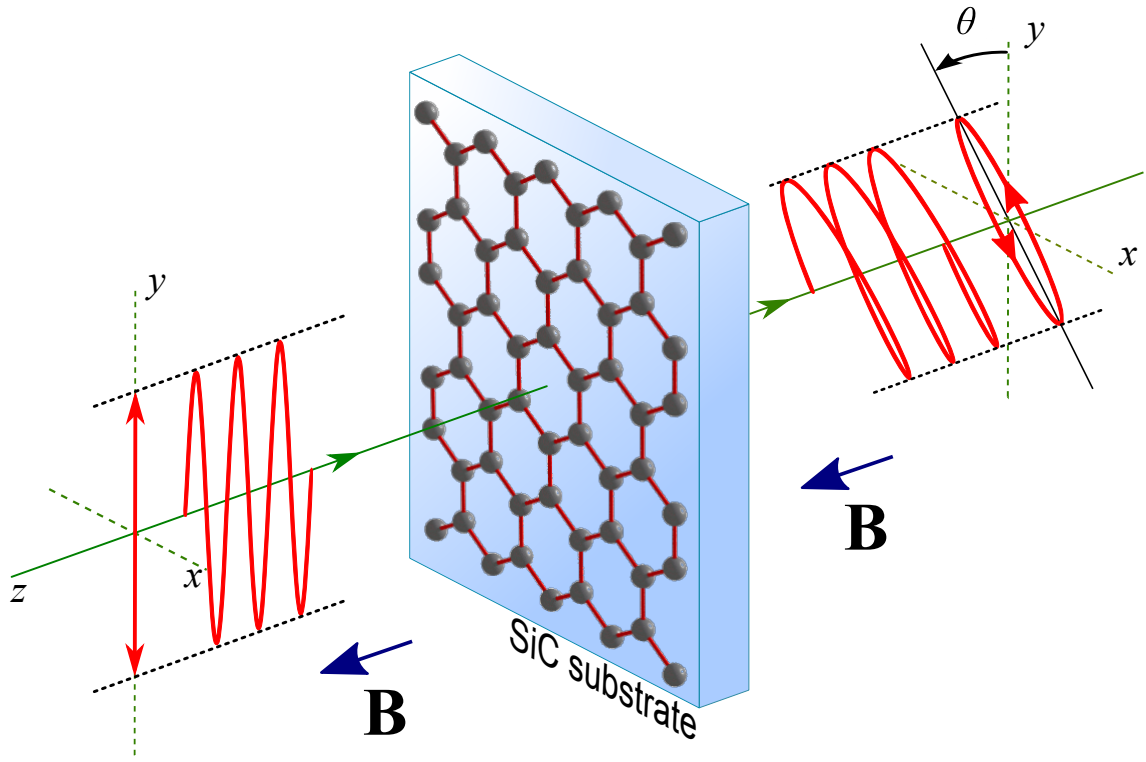


FIG. 1. **Schematics of the magneto-optical Faraday rotation experiment.** The polarization plane of the linearly polarized incoming beam is rotated by the Faraday angle  $\theta$  after passing through graphene on a SiC substrate in a perpendicular magnetic field. Simultaneously, the polarization acquires a certain ellipticity. The positive direction of the magnetic field is along the z-axis.

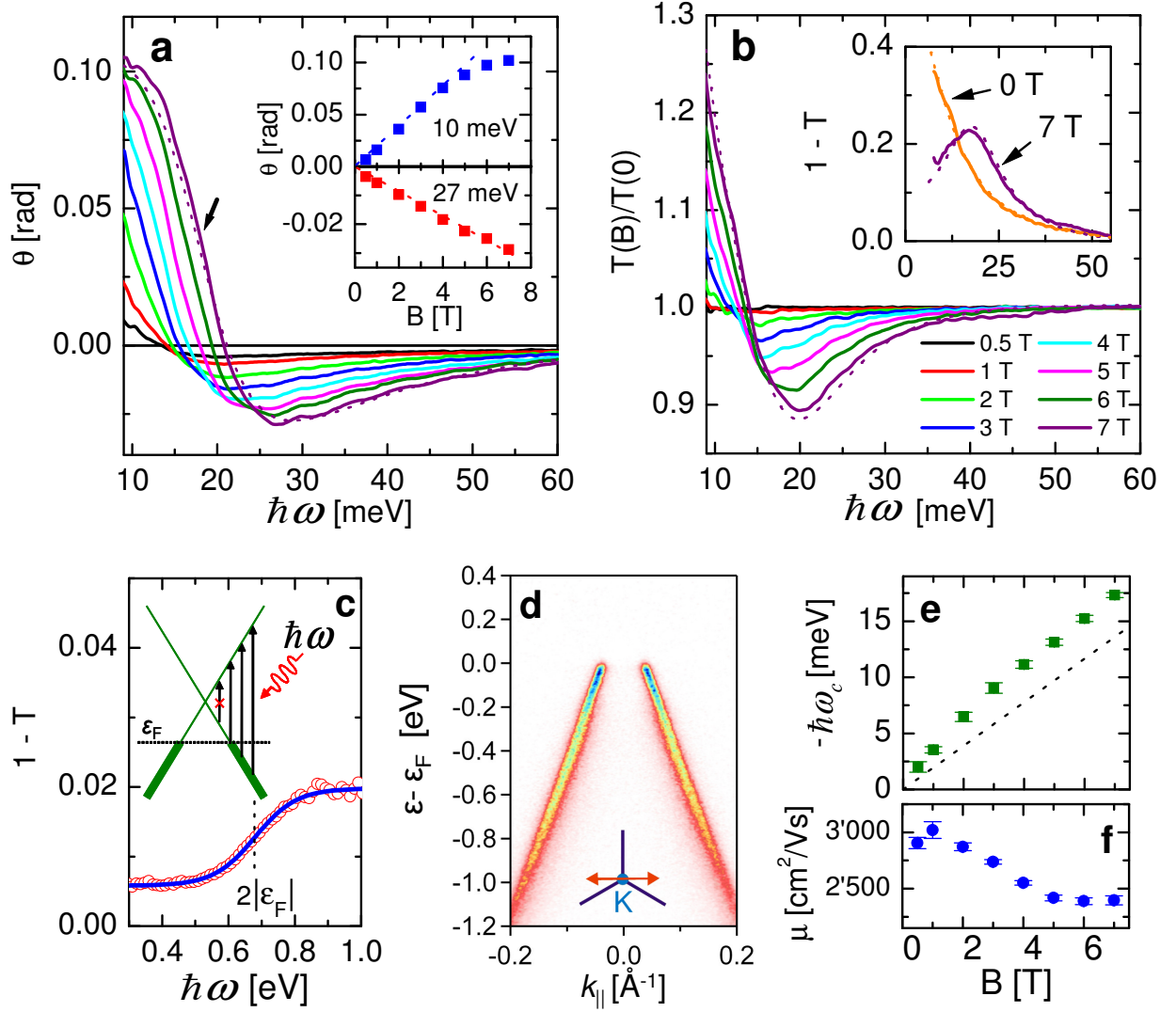


FIG. 2. **Faraday rotation and magneto-optical transmission spectra of single layer graphene.** Panel **a**: the Faraday angle  $\theta$  at several fields up to 7 T at 5 K. The inset presents the magnetic field dependence of  $\theta(B)$  at  $\hbar\omega = 10$  and 27 meV. The dashed lines are linear fits of the data points between 0 and 5 T. Panel **b**: the transmission ratio  $T(B)/T(0)$  at the same fields. In the inset the absorption spectra  $1 - T(B)$  for  $B = 0$  T and 7 T are shown. The dashed lines in both panels and the inset of panel **b** are fits using the classical cyclotron resonance equations (4) and (5). Panel **c**: the absorption of the same sample at room temperature in zero field (symbols), showing a clear step at  $\hbar\omega = 2|\epsilon_F|$  due to Pauli blocking as shown in the inset. The blue line is a fitting curve using a phenomenological broadened step function  $a + b \tanh[(\hbar\omega - 2|\epsilon_F|)/\delta\epsilon]$  giving  $|\epsilon_F| = 0.34 \pm 0.01$  eV. Panel **d**: the band dispersion near the K-point measured by ARPES on another sample prepared in the same way. Panel **e** shows the cyclotron energy as a function of  $B$ . The dashed line is a theoretical dependence based on equation (6) using  $v_F = 1.02 \times 10^6$  and  $\epsilon_F = -0.34$  eV. Panel **f** - the field dependence of the carrier mobility.

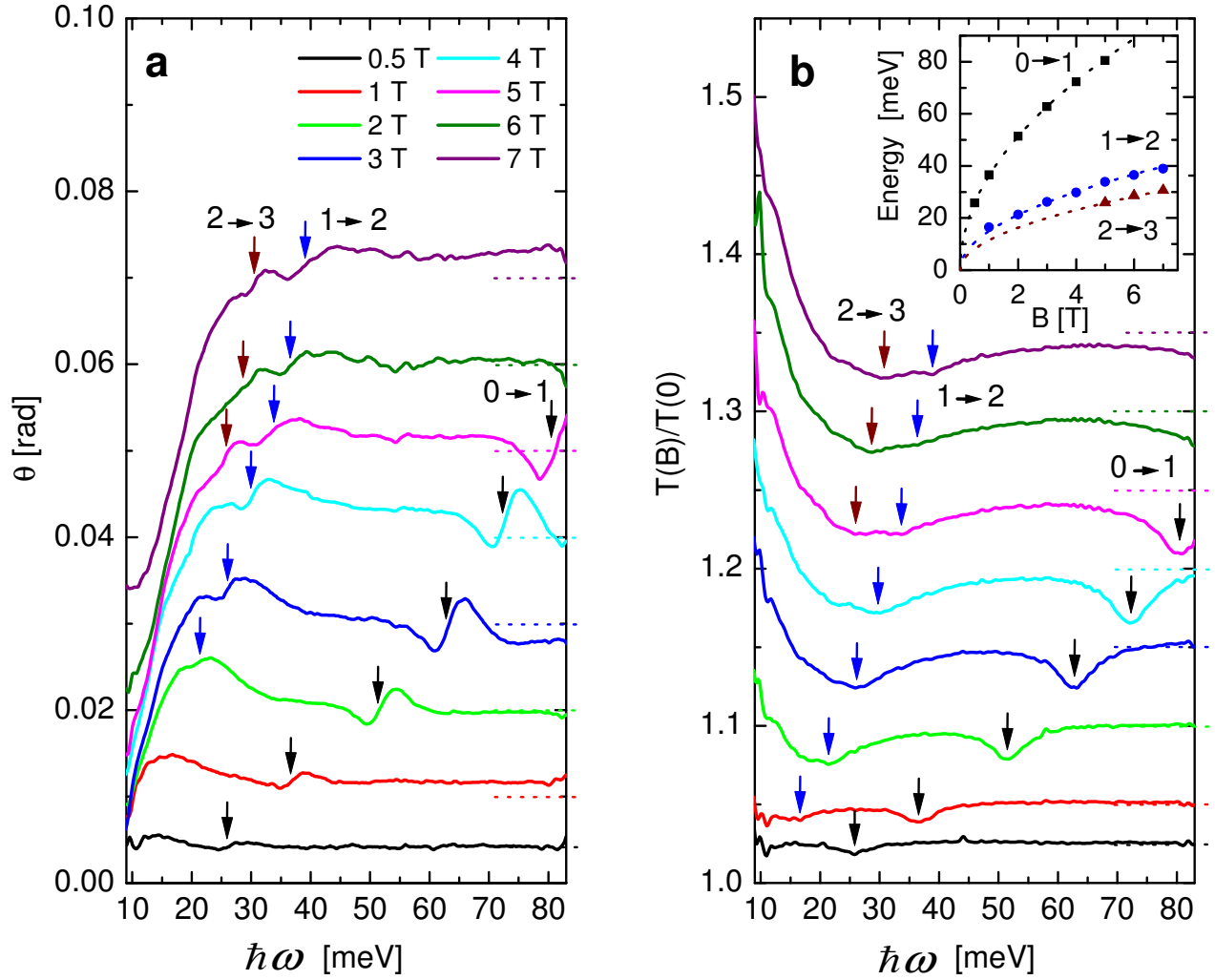


FIG. 3. **Faraday rotation and magneto-optical transmission spectra of multi-layer graphene.** The Faraday angle  $\theta$  (panel **a**) and the transmission ratio  $T(B)/T(0)$  (panel **b**) at several fields up to 7 T at 5 K. The curves on both panels are displaced for clarity; the offsets are given by the dashed lines of the same color. The energies of the inter-LL transitions  $0 \rightarrow 1$ ,  $1 \rightarrow 2$  and  $2 \rightarrow 3$  are indicated by the black, blue and dark red arrows respectively and shown as function of magnetic field in the inset of panel **b**, where also theoretical curves obtained using equation (1) with  $v_F = 1.00 \times 10^6$  m/s are shown as dashed lines.

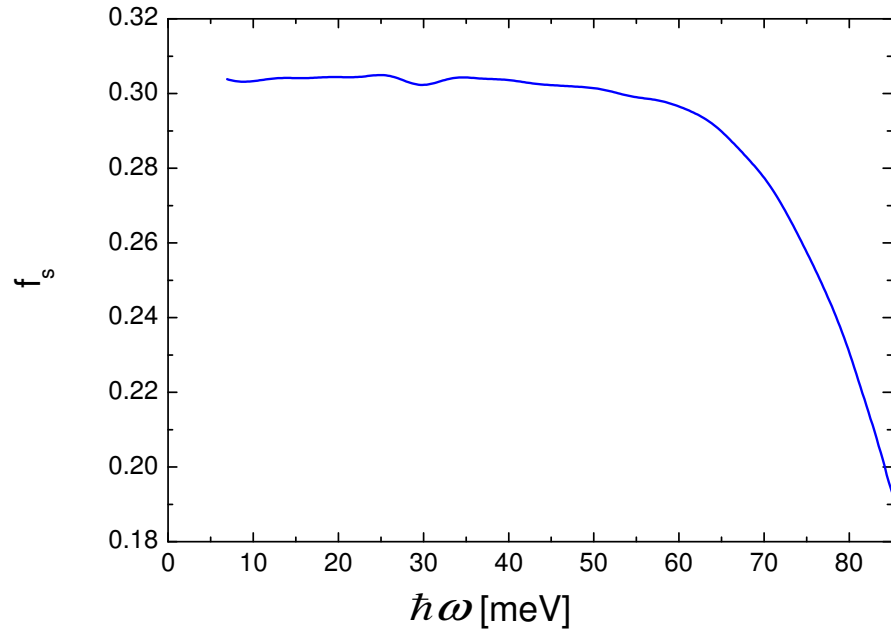


FIG. 4. The function  $f_s(\omega)$  used in the formulas (2) and (3).